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Gold nanotube: structure and melting

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Abstract

In the process of molecular dynamics simulation studies of gold nanowires an interesting structure is discovered. This is a finite double-wall nanowire with a large empty core similar to single-wall and double-wall carbon nanotubes. The structure of the $16-10$ gold nanotube is studied at the room temperature. An investigation of the high-temperature stability has also been carried out. An unusual inward evaporation of atoms from cylindrical liquid walls is found at $T \geq 1200$ K.

Keywords: Gold nanowires; Nanotechnology; Molecular dynamics computer simulation; High resolution transmission electron microscopy

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I. INTRODUCTION

Studies of nanoparticles are very important for advances in various fields of technology. Carbon nanotubes are a topic of experimental and theoretical research in an attractive and already well developed area of condensed matter science [1]. Single-wall and multi-wall carbon nanotubes, as well as their bundles, are synthesized and their structural, thermal, vibrational, mechanical, electronic and transport properties are investigated. Cylindrical nanostructures made of other materials are also the subject of research. For example, Si, BN, $SiSe_2$, WS_2, MoS_2, $NiCl_2$, and various metallic nanowires are studied [1].

Metallic nanowires are interesting from fundamental point of view. They are also important for applications in nanomechanical and nanoelectronic devices. Recently an extensive molecular dynamics (MD) study of unsupported finite and infinite gold nanowires has been carried out [2–9]. The systems analyzed in simulations [2–7] are in the range of radii $R = (0.5 - 1.5)$ nm, the lengths of finite nanowires are $L = (4 - 12)$ nm, and the numbers of particles are $N = (300 - 2100)$. It was found that multi-wall nanowires of lasting stability often form. These gold nanostructures consist of coaxial cylindrical sheets and resemble multi-wall carbon nanotubes. Similar structures are imaged by transmission electron microscopy (TEM) [10–13]. The string tension of thinnest gold nanowires found in the TEM

experiment [10] was calculated by a MD simulation method [8]. Changes of physical properties in a transition from helical and multi-wall gold nanostructures toward face-centered cubic (fcc) ones were analyzed by a genetic algorithm and MD simulations [9].

Multi-shell cylindrical nanostructures found in simulations [2–7] are preserved after a long simulation time of 7 ns. Nanowires whose initial diameters are larger than their lenghts evolve toward an icosahedral shape, a well known structure in cluster physics. Vibrational properties of several multi-wall nanowires are investigated by diagonalization of the dynamical matrix [2]. The averaged coordinates of particles from MD simulations are taken as an input. The results show that the maximal frequencies calculated for cylindrical multi-wall nanowires are higher than for the fcc bulk gold lattice. The capacitance of finite nanometer-scale cylindrical capacitors is also calculated and the values of the order of 0.5 aF are found for length scales where multi-wall nanowires appear in simulations [3]. Solid multi-wall structures are stable up to rather high temperatures $T \sim 900$ K [5]. Nanowires melt much below the bulk melting temperature of gold (~ 1350 K for the potential used in simulations). Melting proceeds by simultaneous disordering of all shells. Deformation properties of multi-wall nanowires under axial compressive loading are studied at $T = 300$ K [7]. Several types of deformation are observed, for example large buckling distortions and progressive crushing. It is found that compressed nanowires recover their initial lengths and radii even after large structural deformations. In contrast to the case of carbon nanotubes, in gold nanowires irreversible local atomic rearrangements occur even under small compressions. Multi-wall gold nanowires are able to sustain a large compressive stress and to store mechanical energy. Structural and melting properties of an unusual finite double-wall gold nanowire are described in this work.

II. COMPUTATIONAL METHOD

In a MD simulation method the equations of motion for the system of particles in a required configuration are solved numerically using suitable algorithms. Simulations of gold nanowires are based on a well-tested embedded atom potential [14]. This potential has been shown to accurately reproduce experimental values for a wide range of physical properties of bulk gold, its surfaces and nanoparticles. A time step of 7.14×10^{-15} s is used. The temperature is controlled by rescaling particle velocities. The MD box of a nanotube consists of 540 atoms. The length of the box is $L = 5$ nm, and the radius is 0.5 nm. A finite nanowire is constructed, i.e., the periodic boundary conditions are not used along the wire axis. First, a nanowire with an ideal fcc structure and the (111)-oriented cross section is prepared at $T = 0$ K. This is done by including all atoms whose distance from the wire axis is smaller than a chosen radius. In previous simulations it was found that an (111) initial orientation of cross sections produces better multi-wall structures than (110) and (100) [5]. An ideal sample is first relaxed at $T = 0$ K. Then, an annealing/quenching procedure is applied. The MD box is heated to $T = 1000$ K. The resulting structure is a double-wall nanotube described in this work. Cylindrical gold nanoparticles with the same radius and the lengths of $2L$ and $3L$ are also simulated. Similar double-wall structures with large empty cores are obtained.

III. RESULTS AND DISCUSSION

A. Low-temperature structure

A double-wall nanotube is shown in Fig. 1. The notation $n_1-n_2-n_3...$, where $n_1 > n_2 > n_3...$, was introduced to label coaxial cylindrical shells in multi-wall metallic nanowires [10]. Figure 1(a) presents a $16-10$ nanowire. As shown in Fig. 1(b), the nanotube is terminated by rounded caps similar to capped carbon nanotubes. Figure 2 shows the central fragment of a nanotube which proves that its walls are made of the triangular lattice of gold atoms. The distance between the neighboring atoms is in the range $(0.25-0.29)$ nm. This should be compared with the bulk interatomic spacing of fcc gold, 0.29 nm. In TEM studies of suspended gold nanowires the distance d between dots on the images was measured [10]. These dots represent positions of gold atoms projected on the image plane. It was found that the distance between dots is in the range $d = (0.25-0.3)$ nm for nanowires with diameters from 0.6 nm to 1.5 nm, and lengths from 3 nm to 15 nm. The average value of this distance for 30 nanowires is $d = 0.288$ nm [10]. These TEM studies of gold nanowires have shown that the outer and inner tubes have the difference of the number of atom rows $n_1 - n_2 = 7$. The exception is the finest studied nanowire $7-1$. However, Kondo and Takayanagi have chosen to treat a single atom chain as "0". The simulation presented here shows that a difference in the number of atom rows is 6, as in the nanowire $7-1$. A similarity between a simulated here $16-10$ nanowire and experimental $7-1$ is in their small radii.

B. Melting

The cross section of a nanotube at several temperatures is shown in Figs. 3 and 4. Close to the room temperature it is possible to distinguish atoms in the trajectories plot. At higher temperatures atoms vibrate more strongly, as shown in Fig. 3(a). At $T = 900$ K diffusion in the walls is intensive, but atoms do not move from the walls (see Fig. 3(b)). At $T = 1200$ K, as shown in Fig. 4(a), atoms in the liquid walls vibrate very strongly. Several atoms evaporate into the empty core. Figure 4(b) shows that atoms evaporate into the core, even when the walls begin to disarrange. Cylindrical liquid walls exist before an evaporation starts. The opposite scenario of the high-temperature disordering (not realized for multi-wall gold nanowires) is a transition from a finite solid cylindrical structure to a solid and liquid blob. The internal energy as a function of temperature is shown in Fig. 5(a). As in other simulated small nanowires [5], the jump in $E(T)$ as a clear sign of the first order phase transition in three-dimensional systems is absent. Nanowires whose radii are of the order of 1 nm are close to one-dimensional systems for which the strict phase transitions do not exist [5]. The average mean-square displacement is shown in Fig. 5(b). The particle displacements sharply increase at 900 K. Therefore, in this nanotube melting starts much below the bulk melting temperature.

IV. CONCLUSIONS

Recently an evidence of a single-wall platinum nanotube was reported [15]. This structure was obtained from a suspended platinum nanowire by electron-beam thinning method and

was imaged in ultrahigh-resolution electron microscope. The outer shell of a $13-6$ double-wall platinum nanowire was stripped and inner shell was exposed. The results presented here show the existence of an unsupported double-wall gold nanotube.

Gold nanowires are very promising for applications in nanodevices, for example as interconnections in integrated circuits. Experimental results [10–13], as well as simulations presented here and in Refs. [2–9], show that the smallest gold nanowires are strong and stable even at high temperatures. These nanowires exist in various exotic forms, from monatomic chains to multi-wall cylindrical structures [2–13]. Transport properties of gold nanowires may also make them very useful [16]. In preparation and investigation of metallic nanowires results obtained by molecular dynamics simulations are important.

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FIGURES

FIG. 1. Atomic positions for a nanotube at $T = 300$ K: (a) top view of a cross section after the caps were removed. This is a $16 - 10$ nanowire (atoms from two layers are completely visible), (b) side view.

FIG. 2. Atomic positions for a side view of partial walls at T=300 K. An irregular cut of a nanotube containing atoms from both walls is presented to show a cylindrical triangular lattice.

FIG. 3. The structural properties of a nanotube: (a) $T = 500$ K, (b) $T = 900$ K. These top views of the central part of a nanotube are represented by the particle trajectory plots and refer to a time span of 3.5 ps. All atoms in the central slice of the thickness of 4 nm along the nanotube axis are included.

FIG. 4. The high-temperature properties of a nanotube: (a) $T = 1200$ K, (b) $T = 1300$ K. Details as in Fig. 3.

FIG. 5. The temperature dependence of: (a) total energy per atom, (b) mean-square displacements.

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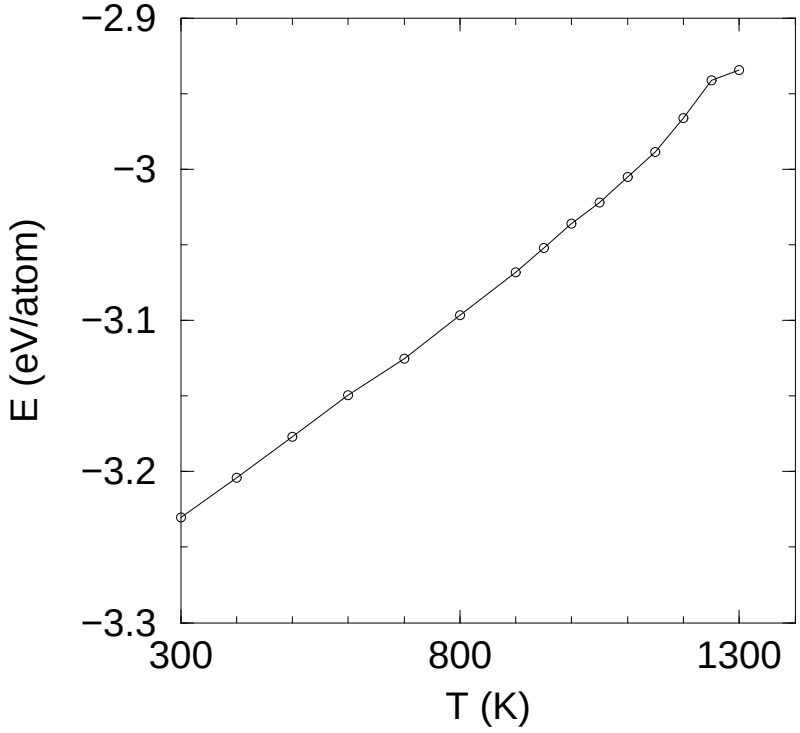
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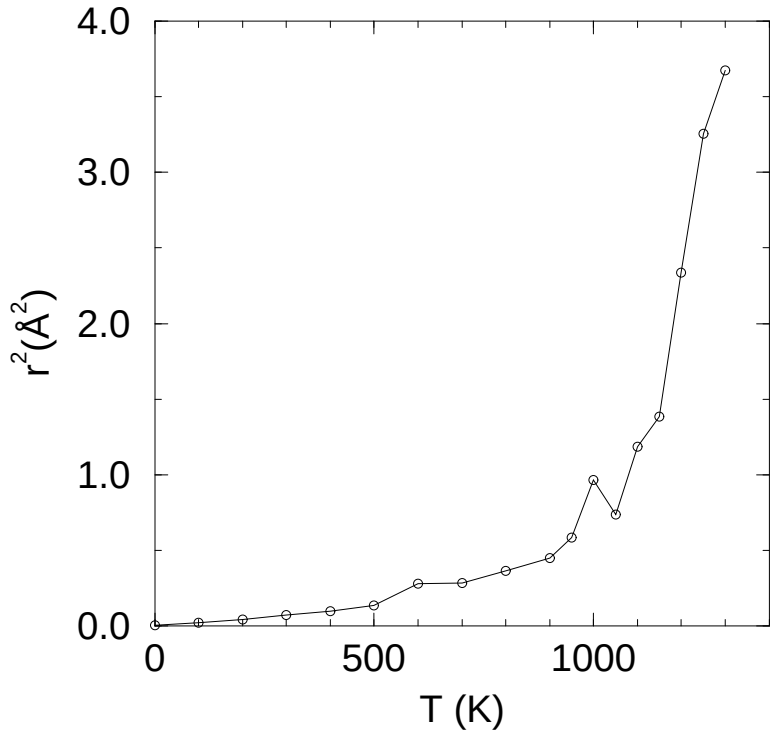
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Fig. 5